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In re Patent Application of

Hisashi KAWANABE, et al.

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FOR: URETHANE RESIN COMPOSITION
FOR OPTOELECTRIC CONVERSION
ELEMENT SEALER

DECLARATION UNDER 37 C.F.R. §1.131
(VERIFICATION OF TRANSLATION)

Assistant Commissioner of Patents
Washington, D.C. 20231

Sir:

I, Hiroyuki KITAHASHI, do hereby solemnly and sincerely declare as follows:

1. I am a translator, residing at 12-21, Sekimachi-kita 2-chome, Nerima-ku, Tokyo, Japan.

2. I translated Japanese Patent Application No. 013786/2000 which is the priority document of the above-identified application and certify that the attached translation is a true translation to the best of my knowledge and belief.

3. Further, I am aware that willful false statements and the like are punishable by fine, imprisonment or both under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Declarant's Signature: H. Kitahashi
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[Name of Document] Specification 1

[Name of Document] Drawing 1

[Name of Document] Abstract 1

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[Document Name] Specification

[Title of the Invention] Material for Light-Emitting Element Sealer

[Claims]

[Claim 1] A material for a light-emitting element sealer, which is obtained by the reaction of an isocyanate compound and a polyol under heating.

[Claim 2] The material for a light-emitting element sealer according to claim 1, wherein the isocyanate compound comprises an aliphatic and/or alicyclic polyisocyanate compound.

[Claim 3] The material for a light-emitting element sealer according to claim 1, wherein the isocyanate compound comprises 2,5- and/or 2,6-diisocyanatomethylbicyclo[2.2.1]heptane.

[Detailed Description of the Invention]

[0001]

[Technical Field of the Invention]

The present invention relates to a sealing material for a light-emitting element such as a light-emitting diode (hereinafter abbreviated to LED).

[0002]

[Prior Art]

Heretofore, as the sealing material for a light-emitting element such as an LED, an epoxy resin composition has been mainly used, and the sealing material can be prepared relatively easily by transfer molding or the like of an epoxy resin composition containing an epoxy resin, a curing agent, a curing accelerator and a mold release agent. Illustrative examples of such an epoxy resin composition disclosed in JP-A 11-43586 include epoxy resins such as a bisphenol A-type, bisphenol F-type and bisphenol S-type, novolac-type epoxy

resins such as ortho-cresol and phenol, and alicyclic epoxy resins, which are relatively less, colored. Illustrative examples of the curing agent include acid anhydrides such as hexahydrophthalic anhydride, methylhexahydrophthalic anhydride and tetrahydrophthalic anhydride, novolac-type resins obtained by subjecting each of phenol, cresol, xylenol, resorcinol and the like and formaldehyde to condensation reaction. Furthermore, there is known an epoxy resin composition for sealing a semiconductor device which contains an amine-based curing agent and the like.

[0003]

On the other hand, JP-A 5-32749 discloses a urethane (meth)acrylate, which has developed as a coating agent for an optical fiber, a resin composition containing thereof and cured products thereof. This material can be also used as a sealer for the LED.

[0004]

[Problem to be solved by the Invention]

However, the epoxy resin which is generally used as a sealer for the light-emitting element of the LED has a problem that it is liable to be colored by heat or light, thereby lowering light transmittance, with the result that the performance of the LED deteriorates, and another problem that it does not cure at sealing on the surface of a surface-mountable LED.

[0005]

Furthermore, in the resin composition using a urethane (meth)acrylate, the urethane (meth)acrylate is obtained by first producing a fluorine-containing (meth)acrylate and then reacting the fluorine-containing (meth)acrylate with a polyol and an organic polyisocyanate. However, this composition is prepared by the two steps of the reactions, and hence it is expensive so that it is

industrially disadvantageous as the sealer for the LED.

[0006]

[Means for solving the problem]

The present inventors have extensively investigated to solve these problems and as a result, they have found that a thermosetting urethane resin is capable of comparatively easily sealing an LED, excellent in curing behavior, scarcely colored and economically obtained, leading to the completion of the present invention.

[0007]

That is, the present invention is as follows:

- 1) A material for a light-emitting element sealer, which is obtained by the reaction of an isocyanate compound and a polyol under heating.
- 2) The material for a light-emitting element sealer according to claim 1, wherein the isocyanate compound comprises an aliphatic and/or alicyclic polyisocyanate compound.
- 3) The material for a light-emitting element sealer according to claim 1, wherein the isocyanate compound comprises 2,5- and/or 2,6-diisocyanatomethylbicyclo[2.2.1]heptane.

[0008]

[Mode for Carrying Out the Invention]

The present invention will be hereinafter described in detail. The isocyanate compound of the invention includes aliphatic and/or alicyclic polyisocyanates such as hexamethylene diisocyanate (HDI), 1-isocyanato-3,3,5-trimethyl-5-isocyanatomethylcyclohexane (IPDI), xylylene diisocyanate (XDI), 2,5- and/or 2,6-diisocyanatomethyl bicyclo[2.2.1]heptane (NDBI), hydrogenated xylylene diisocyanate (hydrogenated XDI), hydrogenated

diphenylmethane diisocyanate (hydrogenated MDI), tricyclododecane diisocyanate (TCDI) and their derivatives such as nurates, allophanates, adducts and urethodiones. Of these polyisocyanates, preferred is 2,5- and/or 2,6-diisocyanatomethyl bicyclo[2.2.1]heptane having bicyclo-ring. This compound can be prepared by a process, for example, disclosed in JP-A-3-220167, wherein 2,5- and/or 2,6-diaminomethyl bicyclo[2.2.1]heptane is treated with hydrogen chloride gas in a solvent mixture composed of isoamyl acetate and o-dichlorobenzene to obtain a hydrochloride salt thereof; then carrying out phosgenation by blowing phosgene in an amount of about 2.2 times the theoretical amount; after the completion of the reaction, blowing inert gas to remove phosgene in the system; then removing the solvent and subjecting the residue to fractional distillation under a reduced pressure.

[0009]

The polyols include diols such as glycerin, ethylene glycol, propylene glycol, dipropylene glycol, 1,3-butanediol, 1,4-heptanediol, neopentyl glycol, 1,5-heptanediol, 1,6-hexanediol, 2,2-dimethyl-1,3-propanediol, 3-methyl-1,5-pentanediol, 2-methyl-2,4-pentanediol, 2-ethyl-1,3-hexanediol, 1,3-hydroxy benzene and 1,3-bis(2-hydroxyethoxy)benzene; trimethylolethane, trimethylolpropane, 1,3-hexanediol, 1,4-hexanediol; triols such as 1,2,6-hexanetriol and their addition products with propylene oxide and ethylene oxide; polyols such as pentaerythritol and their addition products with propylene oxide and ethylene oxide, or polyester polyols, polyether polyols and polyacryl polyols; and saccharides such as inositol, xylitol, sorbitol, fructose and glucose. These polyols can be used solely or in combination of two or more.

[0010]

Although not restricted, the sealing material for a light-emitting element according to the present invention is obtained by reacting a polyisocyanate or its derivative with a polyol under heating. The amounts of polyisocyanate and polyol are preferably 0.8 - 1.2 in term of NCO/OH, more preferably 0.9 - 1.1. The reaction is preferably carried out by adding a polyol to an isocyanate compound and then mixed, and the two are miscible with each other. The reaction can be carried out in the presence or absence of a catalyst. No particular restriction is put in the catalyst if it is a urethanized catalyst, it is preferably an organometallic catalyst, more preferably an organotin catalyst. The use amount of the catalyst, if using, is 0.001 - 5 wt.%, preferably 0.01 - 2 wt.%, based on the isocyanate compound. However, it is more preferable not to use the catalyst in producing a LED. The molding temperature is preferably 40 to 220°C, more preferably 60 to 180°C. The molding time is preferably 1 minute to 10 hours, more preferably 1 to 7 hours. Further, after-curing is preferably carried out at 60 to 180°C for 1 to 12 hours.

[0011]

[Examples]

The present invention will be further described with reference to Examples hereinafter. Here, GPC measurement was carried out by using Shodex AD-8025S as column and N,N-dimethylformamide as a mobile phase.

[0012]

Example 1

While 206.2 g (1 mole) of a mixture of 2,5- and 2,6-diisocyanatobicyclo[2.2.1]heptane (hereinafter referred to as NBDI) were stirred at 25°C, 146.2 g (1 mole) of 2-ethyl-1,3-hexanediol were added thereto.

They were stirred and mixed for 10 minutes to be dissolved homogenously. The homogeneous solution was transferred into a mold, allowed to react in an oven heated to 100°C in an inert gas atmosphere for 5 hours and then after-curing at 150°C for 3 hours to obtain a colorless, transparent polyurethane resin. Figures 1 and 2 show the IR chart and the GPC chart of the polyurethane resin, respectively.

[0013]

Example 2

While 131.6 g (0.9 mole) of 2-ethyl-1,3-hexanediol were stirred at 25°C, 6.1 g (0.066 mole) of glycerin were added thereto to obtain 137.7 g of a solution (hereinafter referred as to "A solution"). While 206.2 g (1 mole) of NBDI were stirred at 25°C, 137.7 g of A solution were added thereto. After stirred and mixed for 10 minutes to be dissolved homogenously, they were allowed to react at 100°C for 5 hours and after-cured at 150°C for 3 hours in the same manner as in Example 1 to obtain a colorless, transparent polyurethane resin.

[0014]

Example 3

While 131.6 g (0.9 mole) of 2-ethyl-1,3-hexanediol were stirred at 25°C, 11.6 g (0.1 mole) of 1,4-hexanediol were added thereto to obtain 143.2 g of a solution (hereinafter referred as to "B solution"). While 206.2 g (1 mole) of NBDI were stirred at 25°C, 143.2 g of B solution were added thereto. After stirred and mixed for 10 minutes to be dissolved homogenously, they were allowed to react at 100°C for 5 hours and after-cured at 150°C for 3 hours in the same manner as in Example 1 to obtain a colorless, transparent polyurethane resin.

[0015]

Example 4

While 206.2 g (1 mole) of NBDI were stirred at 25°C, 146.2 g (1 mole) of 2-ethyl-1,3-hexanediol were added thereto and stirred for 10 minutes, thereby obtaining a homogenous solution. The homogenous solution was allowed to react at 100°C for 5 hours and after-cured at 150°C for 3 hours to form a lamp-shaped LED so that a blue-light-emitting LED chip was obtained.

[0016]

Comparative Example 1

The lamp-shaped LED chip prepared in Example 4 and a blue-light-emitting lamp-shaped LED in which a light-emitting element was sealed with a commercially available bisphenol-type epoxy resin were subjected to a continuous application of an electric current of 50 mA for 1000 hours in a thermostatic chamber at a temperature of 25°C and a humidity of 50%. As a result, the lamp-shaped LED of Example 4 showed no coloration, while the commercially available lamp-shaped LED showed yellowing at a part of the light-emitting element.

[0017]

[Effects of the Invention]

The present invention can provide a sealing material for a light-emitting device, which is a resin obtained by reacting under heating a mixture of an isocyanate compound and an aliphatic and/or alicyclic polyol, which is capable of sealing the LED with relatively ease, which is less colored, which has good curability and which is inexpensive.

[Brief Description of the Drawings]

[Fig. 1]

It is an IR chart of a colorless and transparent polyurethane resin obtained by a method of Example 1.

[Fig. 2]

It is a GPC chart of a colorless and transparent polyurethane resin obtained by a method of Example 1.